SCIENCE FOR CERAMIC PRODUCTION

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MULTILAYER CERAMICS BASED ON ALUMINOSILICATE FOR FILTRATION OF VETERINARY PHARMACEUTICALS

S. M. Azarov,[1] T. A. Azarova,[1] A. I. Rat'ko,[1] P. A. Krasochko,[1] and E. S. Zhuravleva[1]

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The results of studying the structure of porous multilayer ceramic filter elements are given and the possibility of their application for refining and sterilizing filtration of an anti-diarrhea energy-enhancing medicine is assessed.

One of the most difficult and time-consuming operations in the production of veterinary pharmaceuticals is separation of suspensions. The separation technology is hampered by the fact that some biological compounds are labile to mechanical, chemical, and thermal effects; therefore, in some cases it is impossible to use the methods of precipitation, evaporation, and centrifuging. Filters based on hollow fibers, polymeric membranes, and ceramic filter elements with a membrane layer of aluminum oxide [1, 2] are currently widely used for separation, concentration, refining, and sterilizing filtration of medicines.

The Institute of General and Inorganic Chemistry of the National Academy of Sciences of Belarus is researching the development of original multilayer composites based on aluminosilicates produced from local minerals to be used as filtering materials. Ceramic filter elements based on aluminosilicates developed at this institute resist a temperature up to 1000°C, do not oxidize in air, and are able to operate in the pH range from 4 to 9.

The purpose of the present study is to analyze the possibility of using the specified multilayer porous filter elements based on aluminosilicates for refining and sterilizing filtration of medicines, in particular an anti-diarrhea energy-enhancing medicine that is currently extensively used in veterinary [3].

The technological scheme of producing substrates for filtering elements included such operations as mixing initial components, isostatic molding, and sintering. The initial material in preparing the substrate was an aluminosilicate material with particle size of 200 – 630 μm.

[1] Institute of General and Inorganic Chemistry of the National Academy of Sciences of Belarus, Minsk, Belarus; S. N. Vyshelesskii Institute of Experimental Veterinary of the National Academy of Science of Belarus, Minsk, Belarus.

By immersing samples based in a specially prepared suspension, powder layers with particle sizes of 32 – 50 and 5 – 10 μm were consecutively deposited on the exterior or inner surface of sintered tubular filter-element bases. After repeated sintering these layers formed a filtering layer of thickness 400 – 600 μm.

To determine the possibility of refining and sterilizing filtration of aluminosilicate materials, a special set was designed (Fig. 1).

Three multilayer filter elements of the "daisy" shape (Fig. 2*a*) with a diameter of 65 and a length of 230 mm were assembled in parallel in a single module. The use of this type of filter elements increases the total surface area of filtration up to 0.26 m^2, whereas for standard cylindrical filter elements of analogous sizes (Fig. 2*b*) the area is only 0.14 m^2.

The liquid to be filtered was poured into a vessel *1*, a tap *2* was opened, and the liquid penetrated into the body of the

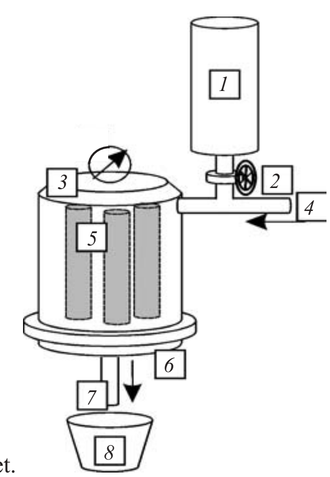

Fig. 1. Scheme of the filtration set.

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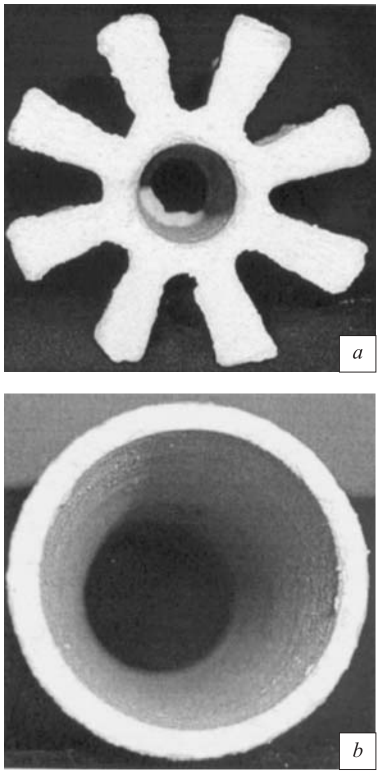

Fig. 2. "Daisy-shaped" filter element (*a*) and cylindrical filter element (*b*).

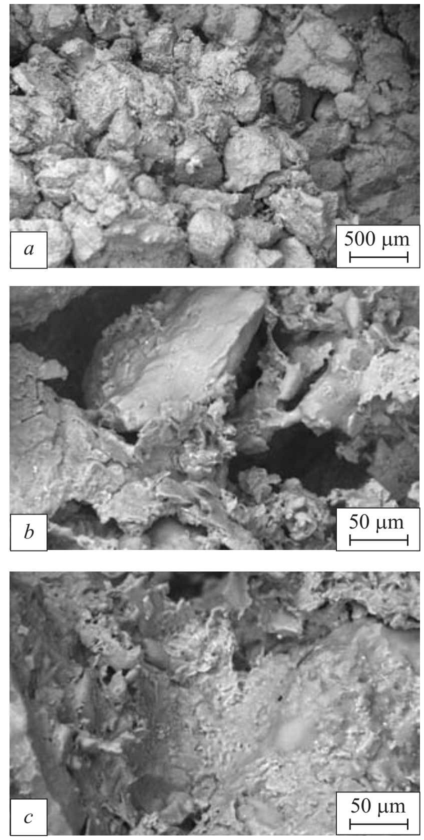

Fig. 3. Material structure of the substrate of a sintered filter element made of aluminosilicate (fracture): *a*) general view (× 35); *b*) pore structure (× 500); *c*) relief of the pore space surface (× 500).

module *3*. After the tap was shut, natural gas was supplied under a pressure via the pipeline *4* and created excessive pressure within the body. This gas was used to push the pharmaceutical liquid through the pores of the filter elements *5* and then the liquid from the collector *6* via the branch line *7* was poured into a recipient vessel *8*. After the end of the purification process the module was washed and sterilized before repeated use.

Before the beginning of the tests with veterinary medicines, the filter elements were tested for their resource on tap water. The resource was estimated based on a decrease in the set efficiency after 7 h of continuous operation under a constant pressure.

The possibility of using the developed porous materials for refining and sterilizing filtration was estimated according to the following technological scheme. The anti-diarrhea energy-enhancing medication was filtered on the previously sterilized set through filter elements in two variants: with a single layer (32 – 50 μm) and with two layers (32 – 50 and 5 – 10 μm). The medicine was regarded as 100% refined when it had no mechanical inclusions of size over 0.5 μm. The sterilizing filtration was performed by repeated transmission of the medicine through the set pretreated in an autoclave.

Information on the material structure of porous multilayer filter elements based on aluminosilicates was obtained using a JSM-5610LV (JEOL) electron microscope.

The material of the substrate of a sintered filter element based on aluminosilicate is shown in Fig. 3. It has a coarse-grained structure with a relatively uniform pore distribution (Fig. 3*a*). The largest and the most extended slot-like pores are located along the grain boundaries.

The configuration of pores and their distribution and size significantly depend on the mechanism of contact formation prevailing in each specific case, which, in turn, is determined by such technological parameters as the powder particle size, the schedule and temperature of sintering, the molding pressure, the batch composition, etc.

The studies performed indicate that the tend toward increasing the total area of the contact surface between the aluminosilicate particles by means of the thermal activation of their interaction in sintering is not justified in filtering materials for fine purification. In this case, along with the growing strength of the filter element (a positive factor), fusing of aluminosilicate particles results in the formation of a coarse-pore material, which has a negative effect on the filtration quality.

Introduction of active additives capable of reacting with each other and with the main component (the aluminosilicate material) makes it possible to obtain pores with an extended structure of a complex configuration (Fig. 3*b*) and thus to in-

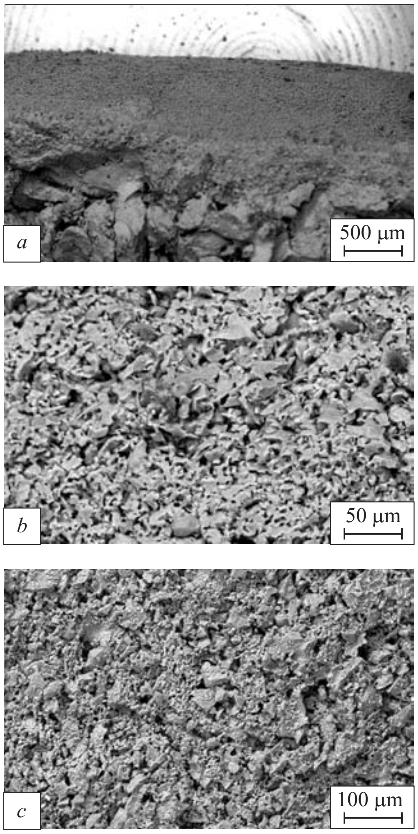

Fig. 4. Structure of the filtering layer of filter element: *a*) filtering layer on the surface of the substrate (radial section, × 35); *b*) fine structure of the filtering layer (radial section, × 500); *c*) structure of the filtering layer (view from above, × 200).

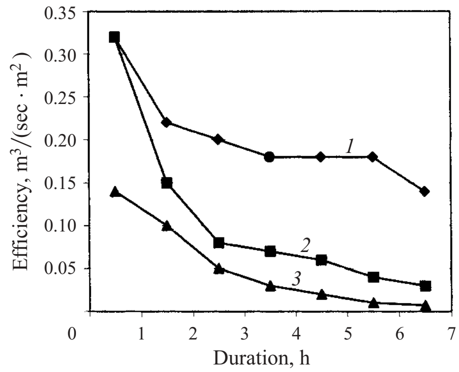

Fig. 5. Dynamics of filtration efficiency loss: *1*) coarse-pore substrate; *2*) filter element with a layer of 32 – 50 μm; *3*) filter element with two consecutively deposited layers of 32 – 50 and 5 – 10 μm.

crease the filtering capacity of the substrate material. Furthermore, the specified technology of producing filter elements based on aluminosilicates creates a surface with an extended relief inside the pore space (Fig. 3*c*), which has a positive effect on the absorption properties of the material.

However, the main service properties of filter elements are determined by the structure of the filtering layer deposited on its surface (Fig. 4*a*). The material of the layer has a cellular finely branched structure with a uniform pore distribution (Fig. 4*b* and *c*), whose average size is 2 – 10 μm.

TABLE 1

Filtering layer		Working pressure, MPa	Efficiency, 10^{-5} $m^3/(sec \cdot m^2)$	Refining, %	Sterilization
type	particles size, μm				
Single-layer	32 – 50	0.050	33.30	95	None
		0.010	6.70	100	None
		0.001	0.16 – 0.33	100	None
Double-layer	32 – 50 and 5 – 10	0.050	25.00	100	None
		0.010	5.00	100	Exists in a limited volume
		0.001	0.16	100	The same

The quality of filtration and the resource of porous materials were evaluated in the course of the study. The efficiency loss dynamics of the materials considered in filtering potable tap water satisfying the Sanitary Norms and Regulations (SanPiN) is shown in Fig. 5. It can be seen that with the given scheme of filtration (the filtration flow is directed from the side of the filtering layer) the resource of the material decreases approximately by half with each consecutive layer deposited. Preliminary experiments in efficiency dynamics for a filtration flow directed from the side of the inner surface of the filter element did not exhibit any significant difference from the dependence represented in Fig. 5.

The results of experiments on refining and sterilizing filtration of the anti-diarrhea energy-enhancing medicine are shown in Table 1.

The studies demonstrated that the multilayer filter elements provide for high-quality refining of biological liquid with a sufficient efficiency within the whole pressure range analyzed. Refining with high efficiency is achieved due to an extended structure and a complex configuration of the pore space of the filtering layers in the materials proposed.

An analysis of the microstructure of the filtering layer found that the pore size (2 – 10 μm against traditionally required 0.2 μm) is insufficiently small for sterilizing filtration. Nevertheless, sterilization was observed in the case of the repeated treatment of up to 100 ml of the liquid with efficiency not greater than 5.0×10^{-5} $m^3/(sec \cdot m^2)$. The partial sterilization observed is due to the sorption of microorganisms on the extended surface. It can be assumed that decreasing the size of pores in the filtering layer to 1.0 – 0.5 μm will subsequently increase the sorption capacity and provide for materials with high sterilizing capacity under working pressures below 0.01 MPa, which will ensure a filtration efficiency of 5.0×10^{-5} $m^3/(sec \cdot m^2)$ for the anti-diarrhea energy-enhancing medicine.

Thus, porous ceramic materials with a multilayer structure are promising for the purification of veterinary medicines.

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